

FORM PTO-1390 (Modified)
(REV 11-98)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

JMYT-235US

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5)

To Be Assigned 09/807682

INTERNATIONAL APPLICATION NO.

PCT/GB99/03269

INTERNATIONAL FILING DATE

04 October 1999 (04.10.99)

PRIORITY DATE CLAIMED

16 October 1998 (16.10.98)

TITLE OF INVENTION

PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE

APPLICANT(S) FOR DO/EO/US

GASCOYNE, John Malcolm; HARDS, Graham Alan; HOGARTH, Martin Philip; and RALPH, Thomas Robertson

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210).
8. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

Items 13 to 20 below concern document(s) or information included:

13. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☒ Certificate of Mailing by Express Mail
20. ☐ Other items or information:

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5) To Be Assigned		INTERNATIONAL APPLICATION NO. PCT/GB99/03269		ATTORNEY'S DOCKET NUMBER JMYT-235US	
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21. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :				CALCULATIONS PTO USE ONLY	
<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO				\$970.00	
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO				\$840.00	
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO				\$690.00	
<input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)				\$670.00	
<input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)				\$96.00	
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$860.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)).				\$0.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	22 - 20 =	2	x \$18.00	\$36.00	
Independent claims	1 - 3 =	0	x \$80.00	\$0.00	
Multiple Dependent Claims (check if applicable).				\$0.00	
TOTAL OF ABOVE CALCULATIONS =				\$896.00	
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable).				\$0.00	
SUBTOTAL =				\$896.00	
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)).				\$0.00	
TOTAL NATIONAL FEE =				\$896.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).				\$40.00	
TOTAL FEES ENCLOSED =				\$936.00	
				Amount to be: refunded	\$
				charged	\$

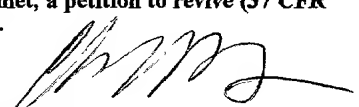
☒ A check in the amount of **\$936.00** to cover the above fees is enclosed.

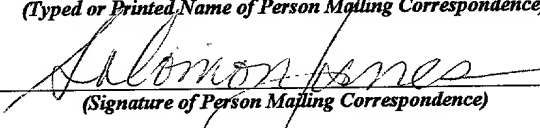
☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees.
 A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **18-0350** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Christopher R. Lewis RATNER & PRESTIA Suite 301 One Westlakes, Berwyn P.O. Box 980 Valley Forge, PA 19482-0980 Phone: 610-407-0700 Fax: 610-407-0701	<div style="text-align: center;">  SIGNATURE </div> <hr/> Christopher R. Lewis NAME
	<hr/> 36,201 REGISTRATION NUMBER
	<hr/> April 16, 2001 DATE

CERTIFICATE OF MAILING BY "EXPRESS MAIL" (37 CFR 1.10)		Docket No. JMYT-235US	
Applicant(s): John Malcolm Gascoyne et al.			
09/807682 Serial No. To Be Assigned	Filing Date Herewith	Examiner	Group Art Unit
Invention: PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE			
<p>I hereby certify that the following correspondence:</p> <div style="border: 1px solid black; padding: 5px; margin: 10px 0;">U.S. National Phase Application w/Form PTO-1390 and all of the documents listed therein</div> <p style="text-align: center;"><i>(Identify type of correspondence)</i></p> <p>is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 in an envelope addressed to: The Assistant Commissioner for Patents, Washington, D.C. 20231</p> <p><u>April 16, 2001</u> <i>(Date)</i></p> <div style="text-align: right; margin-top: 20px;"><p>Solomon James <i>(Typed or Printed Name of Person Mailing Correspondence)</i></p><p> <i>(Signature of Person Mailing Correspondence)</i></p><p>EL751749171US <i>("Express Mail" Mailing Label Number)</i></p></div>			
<p>Note: Each paper must have its own certificate of mailing.</p>			

JMYT-235US

JC03 Rec'd PCT/ETC
PATENT 16 APR 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: John M. Gascoyne et al. : Art Unit:
 Application No.: To Be Assigned : Examiner:
 Filed: Herewith :
 FOR: PROCESS FOR PREPARING A SOLID :
 POLYMER ELECTROLYTE MEMBRANE :

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
 Washington, DC 20231

SIR:

Prior to examination, please amend the above-identified application as follows.

IN THE SPECIFICATION

Please replace the paragraph beginning at page 6, line 23, with the following:

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581 and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO_3H , PO_2H_2 , PO_3H_2 , $\text{CH}_2\text{PO}_3\text{H}_2$, COOH , OSO_3H , OPO_2H_2 , OPO_3H_2 . Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

Please replace the paragraph beginning at page 8, line 8, with the following:

The solid polymer electrolyte membrane prepared by the process of the present invention may be employed as is or as a component of the ion-exchange membrane of a solid polymer electrolyte chemical cell, such as a PEMFC. Accordingly, the present invention further provides a membrane prepared by the process of the present invention. The membrane prepared by the process of the present invention is suitable for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than 200 μ m and preferably less than 100 μ m.

IN THE CLAIMS:

Please replace claims 1-5, 8-14, and 16-22 with the following amended claims.

- 1 1. (Amended) A process for preparing a solid polymer
2 electrolyte membrane comprising an ion-conducting polymer, a catalyst and a high
3 surface area supported material, which process comprises:
 - 4 (a) associating the catalyst with the support material to form a
5 catalysed support; and
 - 6 (b) combining the catalysed support with the ion-conducting
7 polymer.
- 1 2. (Amended) A process according to claim 1, wherein step (b)
2 comprises combining the catalysed support with the ion-conducting polymer in a
3 liquid medium that is aqueous-based and is essentially free from organic solvents.
- 1 3. (Amended) A process according to claim 1, wherein the
2 catalyst comprises one or more precious metals, or combinations thereof, and/or
3 other transition group metals.
- 1 4. (Amended) A process according to claim 1, wherein the
2 catalyst comprises platinum.
- 1 5. (Amended) A process according to claim 1, wherein the
2 catalyst is deposited onto the support material to a loading of between 0.01 to
3 50.0% by weight of the total catalysed support.

1 8. (Amended) A process according to claim 1, wherein the
2 amount of catalysed support incorporated into the membrane is such that the metal
3 loading is lower than 0.1mg/cm².

1 9. (Amended) A process according to claim 8, wherein the
2 amount of catalysed support incorporated into the membrane is such that the metal
3 loading is lower than 0.05mg/cm².

1 10. (Amended) A process according to claim 9, wherein the
2 amount of catalysed support incorporated into the membrane is such that the metal
3 loading is lower than 0.03mg/cm².

1 11. (Amended) A process according to claim 1, wherein the high
2 surface support material is non-electrically conducting.

1 12. (Amended) A process according to claim 1, wherein the high
2 surface area support material is selected from the group consisting of silica, titania,
3 alumina, zirconium oxides, zirconium silicates, tungsten oxides, tin oxides and
4 zeolites.

1 13. (Amended) A process according to claim 1, wherein the
2 support material is in the form of fibres.

1 14. (Amended) A process according to claim 1, wherein the
2 support material is in the form of particles with a mean particle size in the range of
3 from 0.001µm to 10µm.

1 16. (Amended) A process according to claim 1, wherein the ion-
2 conducting polymer comprises an essentially aqueous solution of a perfluorinated
3 co-polymer with ion-exchange groups.

1 17. (Amended) A process according to claim 1, wherein the
2 catalysed support is in particle or fibre form and step (b) comprises directly adding
3 the catalysed support to a solution of the ion-conducting polymer electrolyte.

1 18. (Amended) A process according to claim 1, wherein the
2 catalysed support is in particle form and is applied as a binder to form a fibre

3 network to which the ion-conducting polymer is subsequently applied to produce the
4 membrane.


1 19. (Amended) A process according to claim 1, wherein the
2 catalysed support is in fibre form and itself is formed into a fibre network which is
3 thereafter bound with a binder, and the ion-conducting polymer is subsequently
4 applied to produce the membrane.

1 20. (Amended) A membrane prepared by a process according to
2 claim 1.

1 21. (Amended) A membrane electrode assembly comprising a
2 membrane prepared by a process according to claim 1.

1 22. (Amended) A fuel cell comprising a membrane prepared by a
2 process according to claim 1.

Respectfully submitted,



Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb

Dated: April 16, 2001

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Date of Deposit: April 16, 2001

I hereby certify that this paper and fee are being deposited, under 37 C.F.R. § 1.10 and with sufficient postage, using the "Express Mail Post Office to Addressee" service of the United States Postal Service on the date indicated above and that the deposit is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.

The Assistant Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. 18-0350 of any fees associated with this communication.



Solomon James

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

Specification at page 6, line 24:

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581[, WO 95/08581] and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO_3H , PO_2H_2 , PO_3H_2 , $\text{CH}_2\text{PO}_3\text{H}_2$, COOH , OSO_3H , OPO_2H_2 , OPO_3H_2 . Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

Specification at page 8, line 8:

The solid polymer electrolyte membrane prepared by the process of the present invention may be employed as is or as a component of the ion-exchange membrane of a solid polymer electrolyte chemical cell, such as a PEMFC. Accordingly, the present invention further provides a membrane prepared by the process of the present invention. The membrane prepared by the process of the present invention is [suitably] suitable for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than $200\mu\text{m}$ and preferably less than $100\mu\text{m}$.

IN THE CLAIMS:

- 1 1. (Amended) A process for preparing a solid polymer
2 electrolyte membrane comprising an ion-conducting polymer, a catalyst and a high
3 surface area supported material, which process comprises:
- 4 (a) associating the catalyst with the support material to form a
5 catalysed support; and
- 6 (b) combining the catalysed support with [an] the ion-conducting
7 polymer [composition].
- 1 2. (Amended) A process according to claim 1, [which
2 comprises, as] wherein step (b)[,] comprises combining the catalysed support with
3 [an] the ion-conducting polymer in a liquid medium that is aqueous-based and is
4 essentially free from organic solvents.
- 1 3. (Amended) A process according to claim 1 [or claim 2],
2 wherein the catalyst comprises one or more precious metals, or combinations
3 thereof, and/or other transition group metals.
- 1 4. (Amended) A process according to [any preceding] claim 1,
2 wherein the catalyst comprises platinum.
- 1 5. (Amended) A process according to [any preceding] claim 1,
2 wherein the catalyst is deposited onto the support material to a loading of between
3 0.01 to 50.0% by weight of the total catalysed support.
- 1 8. (Amended) A process according to [any preceding] claim 1,
2 wherein the amount of catalysed support incorporated into the membrane is such
3 that the metal loading is lower than 0.1mg/cm².
- 1 9. (Amended) A process according to claim 8, wherein the
2 amount of catalysed support incorporated into the membrane is such that the metal
3 loading is lower than 0.05mg/cm².

1 10. (Amended) A process according to claim 9, wherein the
2 amount of catalysed support incorporated into the membrane is such that the metal
3 loading is lower than $0.03\text{mg}/\text{cm}^2$.

1 11. (Amended) A process according to [any preceding] claim 1,
2 wherein the high surface support material is non-electrically conducting.

1 12. (Amended) A process according to [any preceding] claim 1,
2 wherein the high surface area support material is selected from the group consisting
3 of silica, titania, alumina, zirconium oxides, zirconium silicates, tungsten oxides,
4 tin oxides and zeolites.

1 13. (Amended) A process according to [any preceding] claim 1,
2 wherein the support material is in the form of fibres.

1 14. (Amended) A process according to [any one of claims 1 to
2 12] claim 1, wherein the support material is in the form of particles with a mean
3 particle size in the range of from $0.001\mu\text{m}$ to $10\mu\text{m}$.

1 16. (Amended) A process according to [any preceding] claim 1,
2 wherein the ion-conducting polymer [composition is] comprises an essentially
3 aqueous solution of a perfluorinated co-polymer with ion-exchange groups.

1 17. (Amended) A process according to [any preceding] claim 1,
2 wherein the catalysed support[, is in particle or fibre form[, is added] and step (b)
3 comprises directly adding the catalysed support to a solution of the ion-conducting
4 polymer electrolyte.

1 18. (Amended) A process according to [any of claims] claim 1
2 [to 11 or 13 to 15], wherein the catalysed support is in particle form and is applied
3 as a binder to form a fibre network to which the ion-conducting polymer is
4 subsequently applied to produce the membrane.

1 19. (Amended) A process according to [any of claims] claim 1
2 [to 13 or 16], wherein the catalysed support is in fibre form and itself is formed
3 into a fibre network which is thereafter bound with a binder, and the ion-conducting
4 polymer is subsequently applied to produce the membrane.

1 20. (Amended) A membrane prepared by a process according to
2 [any preceding] claim 1.

1 21. (Amended) A membrane electrode assembly comprising a
2 membrane prepared by a process according to [any one of claims] claim 1 [to 19].

1 22. (Amended) A fuel cell comprising a membrane prepared by a
2 process according to [any one of claims] claim 1 [to 19].

PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE

The present invention relates to a process for the preparation of a solid polymer electrolyte membrane that is of use in electrochemical devices, particularly fuel cells.

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Electrochemical cells invariably comprise an ion-conducting electrolyte located between two electrodes, the anode and cathode, at which the desired electrochemical reactions take place. Electrochemical cells may be found in a range of devices, for example fuel cells, batteries, sensors, electrodialysis reactors and electrolytic reactors. They have a diverse range of applications, including the electrolysis of water, chemical synthesis, salt splitting, water purification, effluent treatment and metal finishing, among others.

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A fuel cell is an energy conversion device that efficiently converts the stored chemical energy of its fuel and oxidant into electrical energy. It does so by combining either hydrogen, stored as a gas or methanol, stored as a liquid or a gas, with oxygen to generate electrical power. The hydrogen or methanol is oxidised at the anode and oxygen is reduced at the cathode. Both electrodes are of the gas diffusion type. The combined structure formed from the membrane and the two gas diffusion electrodes is known as the membrane electrode assembly (MEA). An electric current is produced by the electron transfer reaction on the electrodes, with the flow of ions in the electrolyte completing the electrical circuit between the electrodes. The electrolyte therefore has to be in contact with both electrodes, and may be acidic or alkaline, and liquid or solid, in nature. In proton exchange membrane fuel cells (PEMFC), the electrolyte is a solid, ion-conducting, *i.e.* a proton-conducting, polymer membrane. The movement of ions between the electrodes is dependent upon the presence of water in the membrane. The membrane must be fully humidified to provide the efficient movement of ions *i.e.* maintain the highest possible specific conductivity. The membrane also prevents large quantities of hydrogen gas supplied at the anode and oxygen supplied at the cathode from passing through the electrolyte and mixing, although there is some transport of the reactant gases - a phenomenon referred to as "cross-over" - which leads to a reduction in cell performance. It is important that this cross-over is minimised. This is a particular problem with conventionally employed membranes at thicknesses below around 40µm, which are otherwise favoured because of their high conductivity in the fuel cell.

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Conventionally, the membrane electrolytes useful in fuel cells are selected from commercially available membranes, for example the perfluorinated membranes sold under the trade names Nafion® (E I DuPont de Nemours and Co), Aciplex® (Asahi Chemical Industry) and Flemion® (Asahi Glass KK). In PEMFCs, the membrane electrolyte films - such as the Nafion® types of DuPont - have to be maintained in a highly water-saturated form in order to maximise their proton conductivity properties. Maintaining this high level of water content under operating conditions requires a complex water management system.

Attempts to control the humidity of the membrane have been made, which include saturating the hydrogen gas supplied to the anode and air supplied to the cathode with water vapour. However, this method brings with it its own disadvantages: it makes the overall system more complicated; lowers the fuel cell system efficiency; and can also reduce the cell's performance, particularly at high current densities, due to increased reactant gas (particularly oxygen in air) diffusional problems as a result of the increased amount of water in the electrode layers.

It has been recognised that one way to overcome these difficulties is to employ the concept of a self-humidified membrane. One way that has been suggested that this can be achieved is through the use of water-retaining particles such as highly-dispersed silica (SiO₂) incorporated into the membrane electrolyte film. The membrane can retain a higher level of water and is able to function with reduced need for humidifying the inlet gases.

It has also been discovered (European patent application EP 0631337) that it is possible to incorporate catalyst materials, such as those based on platinum, into the particle containing membrane. This further improves the water retaining ability of the membrane by reacting any hydrogen and oxygen that diffuse into the membrane from each of the catalysed electrode layers to produce water *in situ*. This also can permit the use of thinner membranes, with higher conductivity, by reducing the deleterious effects of reactant gas cross-over that would otherwise occur. Furthermore, the ability to use thinner membranes itself will further reduce the requirements for external humidification of the reactant gases. The back diffusion of water from cathode to anode is enhanced, thereby preventing the anode face of the membrane from drying.

The disclosure of EP 0631337 relates to a solid polymer electrolyte composition comprising solid polymer electrolyte and 0.01 to 80% (based on weight of electrolyte) of at least one metal catalyst, and the use of this composition in fuel cells. The specification further discloses the optional incorporation of 0.01 to 50% of particles and/or fibres of one or more metal oxides into the electrolyte. However, the method disclosed for making such compositions is multi-step and also has associated disadvantages. For example, one such composition is prepared by mixing a 5% w/w isopropanol solution of the electrolyte and an isopropanol dispersion of particles and/or fibres of 0.01 to 50% silica or titania and sufficiently agitating the mixture in an ultrasonic homogeniser. After this, the solution is transferred into a membrane moulding vessel and dried at room temperature, then vacuum-dried at 60°C to remove the isopropanol and form the membrane. To incorporate the metal catalyst, in the case of platinum, the solid polymer electrolyte membrane containing the metal oxide is dipped in distilled water containing a platinum-ammine complex, followed by agitation at 60°C for 5 hours, during which the platinum complex replaces the protons in the membrane by ion-exchange. After washing the membrane in distilled water for over 4 hours, it is dipped in distilled water containing a twenty times equivalence of hydrazine as reductant. After agitation at 60°C for another 5 hours the platinum particles are deposited in the solid polymer electrolyte containing the metal oxide. Then, after the ion-exchange group is protonised by means of treatment with 4 molar concentration hydrochloric acid, the membrane is washed and dried.

Hence, it can be seen that the prior art process for forming the material is complex and involves multiple steps, many of which are time-consuming and may not be easily capable of producing a consistent material. These factors increase the cost of producing this self-humidified membrane. It is essential for the future commercialisation of PEMFC power generation systems that the cost of the materials employed should be minimised. The polymer electrolyte membrane is one of the most expensive materials employed in the PEM fuel cell stack, due principally to the nature of the polymer itself, and its cost is currently too high to enable widespread commercialisation. Therefore, any process for producing more complex membranes needs to be simple, straightforward and low-cost.

A further expensive material in the PEM fuel cell stack is the platinum catalyst conventionally employed in the anode and cathode catalyst layers to promote the fuel cell electrode reactions of hydrogen oxidation and oxygen reduction. For many applications, particularly transportation, there is a need to minimise the use of these materials. It is widely
5 believed that, for successful commercial exploitation, the level of platinum in the fuel cell stack should be reduced to a cell loading of as low as 0.20mg/cm^2 of the MEA. Therefore, whilst the incorporation of an additional amount of platinum catalyst into the membrane to effect the combination of any reactant gases permeating into the membrane to produce an improved self-humidified membrane is a worthwhile objective, it is a requirement that this
10 additional amount of platinum be as low as possible. EP 0631337 and J. Electrochem. Soc. 143 (12) 3847-52 (1996) indicate that platinum loadings of 0.07mg/cm^2 are employed in the films produced by the above described multi-step process. At some 35% of the total platinum content that may be allowable for vehicular applications of the PEM fuel cell, this may be too high a loading for economic reasons. To our knowledge, therefore, membranes of this type
15 have not yet found application in any of the current, practical pre-production demonstrations of PEM fuel cell power generators.

It is therefore an object of the present invention to overcome the disadvantages of the conventional ion-exchange membranes, without incurring the disadvantages of the prior art
20 process for manufacturing a self-humidifying membrane.

Accordingly, the present invention provides a process for preparing a solid polymer electrolyte membrane comprising an ion-conducting polymer, a catalyst and a high surface area support material, which process comprises:

- 25 (a) associating the catalyst with the support material to form a catalysed support; and
(b) combining the catalysed support with an ion-conducting polymer composition.

Preferably, the ion-conducting polymer composition is an aqueous ion-conducting polymer composition, for example as disclosed in European patent specification number EP
30 0731520, the contents of which is incorporated herein by reference in their entirety. Accordingly, the process of the present invention preferably comprises, as step (b), combining

the catalysed particulate material with an ion-conducting polymer in a liquid medium that is aqueous-based and is essentially free from organic solvents.

The process according to this invention, compared with the prior art multi-step process, is less complex, has fewer and shorter process steps, and is capable of producing a consistent and reproducible product; hence, the cost of producing the membrane is thereby reduced. Furthermore, as the catalyst is directly applied to the high surface area particulate material, which thereby acts as the support for the catalyst, the catalyst is also deposited in a high surface area, small particle size form. This enables the expensive catalyst to be used more effectively and in lower amounts whilst also improving the self-humidifying performance of the membrane. Additionally, when the particulate material has water-retaining properties, all of the catalyst promoting the recombination of hydrogen and oxygen is in immediate and intimate contact with the sites at which the water formed can be stored.

The catalyst may comprise any catalyst material capable of enabling the reaction of hydrogen and oxygen to form water to occur at PEM fuel cell temperatures of typically below 150°C. These include the precious metals, platinum, palladium or rhodium, or combinations thereof, or other transition group metals such as nickel or cobalt. Preferably, the precious metals platinum, palladium or rhodium, or combinations thereof are employed and, more preferably, platinum is used. Suitable catalysation processes, as already practised on a commercial scale, can be employed to form the catalyst material. These processes permit very close control of the amount of catalyst metal deposited onto the support, and thus subsequently incorporated into the membrane. Typically the catalyst metal can be deposited onto the support to loadings of between 0.01 to 50.0% by weight of the total catalysed support, and more preferably from 1 to 25 wt%, such as, for example 1-10 wt%. The amount of catalysed support incorporated into the membrane is such that the metal loading on the membrane is lower than 0.1mg/cm², preferably lower than 0.05mg/cm² and, more preferably, lower than 0.03mg/cm².

The high surface area support material may comprise any known, non-electrically conducting material which can be used as a support for a catalyst, such as silica, titania, alumina, zirconium oxides, zirconium silicates, tungsten oxides, tin oxides and zeolites. It may also be

advantageous to employ support materials that are known to retain high levels of water once suitably wetted, such as, for example, forms of silica. The material may be in the form of either fibres or small particles, such as particles with a mean particle size in the range of from 0.001 μ m to 10 μ m, preferably 0.01 μ m to 5 μ m.

5

For PEM fuel cell applications, the ion-conducting polymer is a proton-conducting polymer, examples of such polymers being well known to those skilled in the art. More than one proton-conducting polymer may be present and/or a non-ion-conducting polymer may also be included in the membrane prepared by the process of the invention.

10

The proton conducting polymers suitable for use in the present invention may include, but are not limited to:

1) Polymers which have structures with a substantially fluorinated carbon chain optionally having attached to it side chains that are substantially fluorinated. These polymers contain sulphonic acid groups or derivatives of sulphonic acid groups, carboxylic acid groups or derivatives of carboxylic acid groups, phosphonic acid groups or derivatives of phosphonic acid groups, phosphoric acid groups or derivatives of phosphoric acid groups and/or mixtures of these groups. Perfluorinated polymers include Nafion[®], Flemion[®] and Aciplex[®] commercially available from E. I. DuPont de Nemours (U.S. Patents 3,282,875; 4,329,435; 4,330,654; 4,358,545; 4,417,969; 4,610,762; 4,433,082 and 5,094,995), Asahi Glass KK and Asahi Chemical Industry respectively. Other polymers include those covered in U.S. Patent 5,595,676 (Imperial Chemical Industries plc) and U.S. Patent 4,940,525 (Dow Chemical Co.)

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581, WO 95/08581 and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO₃H, PO₂H₂, PO₃H₂, CH₂PO₃H₂, COOH, OSO₃H, OPO₂H₂, OPO₃H₂. Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

3) Fluorinated polymers such as those disclosed in EP 0 331 321 and EP 0345 964

(Imperial Chemical Industries plc) containing a polymeric chain with pendant saturated cyclic groups and at least one ion exchange group which is linked to the polymeric chain through the cyclic group.

4) Aromatic polymers such as those disclosed in EP 0 574 791 and US Patent 5,438,082 (Hoechst AG) for example sulphonated polyaryletherketone. Also aromatic polymers such as polyether sulphones which can be chemically grafted with a polymer with ion exchange functionality such as those disclosed in WO 94/16002 (Allied Signal Inc.).

5) Nonfluorinated polymers include those disclosed in U.S. Patent 5,468,574 (Dais Corporation) for example hydrocarbons such as styrene-(ethylene-butylene)-styrene, styrene-(ethylene-propylene)-styrene and acrylonitrile-butadiene-styrene co- and terpolymers where the styrene components are functionalised with sulphonate, phosphoric and/or phosphonic groups.

6) Nitrogen containing polymers including those disclosed in U.S. Patent 5,599,639 (Hoechst Celanese Corporation), for example, polybenzimidazole alkyl sulphonic acid and polybenzimidazole alkyl or aryl phosphonate.

7) Any of the above polymers which have the ion exchange group replaced with a sulphonyl chloride (SO_2Cl) or sulphonyl fluoride (SO_2F) group rendering the polymers melt processable. The sulphonyl fluoride polymers may form part of the precursors to the ion exchange membrane or may be arrived at by subsequent modification of the ion exchange membrane. The sulphonyl halide moieties can be converted to a sulphonic acid using conventional techniques such as, for example, hydrolysis.

Non-ion conducting polymeric materials which may be used in addition to the one or more ion conducting or proton conducting polymers include PTFE, FEP, PVDF, Viton® and hydrocarbon types such as polyethylene, polypropylene and polymethylmethacralate.

Other ion-conducting polymeric materials which are not proton conducting polymers may be used. For example, such polymers can be used for applications requiring an anion exchange membrane. Anion exchange polymers are generally based on quaternary ammonium groups, rather than the fixed sulphonic acid groups in proton conducting polymers. These include, for example, the tetraalkyl ammonium group ($-\text{N}^+\text{R}_3$) and the quaternary ammonium centre in Tosflex® membranes ($-\text{N}(\text{R}_1)(\text{CH}_2)_y\text{N}^+(\text{R}_3)$) supplied by Tosoh. However, it can be

envisaged that all of the proton exchange polymers described above could have anion exchange equivalents.

Most preferably, the ion-conducting polymer is an essentially aqueous solution of a
5 perfluorinated co-polymer with ion-exchange groups, such as Nafion[®] (as mentioned hereinbefore).

The solid polymer electrolyte membrane prepared by the process of the present invention may be employed as or as a component of the ion-exchange membrane of a solid
10 polymer electrolyte chemical cell, such as a PEMFC. Accordingly, the present invention further provides a membrane prepared by the process of the present invention. The membrane prepared by the process of the present invention is suitably for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than 200µm and preferably less than 100µm.

15

The catalysed support can be incorporated into the membrane by a number of methods:

The catalysed support, when in particle or fibre form, can be added directly to an
20 solution of the proton-conducting polymer electrolyte material; preferably, an essentially aqueous solution as described in EP 0731520; the use of an aqueous solution permits the direct addition of the pre-formed catalysed support to the polymer solution, and eliminates the undesirable risk of combustion of the organic solvents that are present in the typically-employed polymer solutions. It is then possible to cast a membrane directly from this mixture
25 according to any of the commonly practised methods in the art to produce a conventional unreinforced membrane. Alternatively, the mixture can be used to form a composite membrane by applying it to a pre-formed reinforcing substrate material such as disclosed in the art, for example an expanded PTFE matrix as disclosed in US 5,599,614 (Bahar et al) or a woven or non-woven fibre network of, for example, glass, silica or polymer fibres as disclosed for
30 example in EP 0875524, unpublished UK Patent Application No 9822569.1 or unpublished UK Patent Application No 9822571.7.

Alternatively, when used in a composite membrane, the catalysed support may, when in particle form, itself comprise the binder used to form the fibre network. Subsequent to the formation of the fibre network or matrix comprising the catalysed support as a binder, a proton-conducting polymer is coated onto the network to produce the final membrane.

- 5 Alternatively, when in fibre form, the catalysed support may itself form the fibre network prior to coating with a binder and subsequently coating with proton-conducting polymer to form the membrane.

- 10 In a further embodiment, a laminated membrane comprising more than one polymer-containing layer is provided, at least one layer of which is a membrane prepared by the process of the invention. Using such a laminated structure, it is possible, for example, to tailor the properties of the laminate membrane opposed to the anode and cathode sides in the MEA of a proton exchange membrane fuel cell, for example, to improve water management in the fuel cell, or to be able to use lower cost proton-conducting polymers to form a substantial part of the laminate membrane.

- 15 The present invention also relates to an MEA and a method for the manufacture thereof, wherein the MEA comprises a membrane produced by a process according to the present invention. A further aspect of the present invention relates to a fuel cell and a method for the manufacture thereof, which fuel cell comprises such a membrane and/or MEA.

The present invention will now be described by way of the following non-limiting example(s).

25 EXAMPLE 1

Catalyst Preparation

- 30 A 5.6 wt% Pt catalyst supported on silica (Davisil™, grade 644 from Aldrich, The Old Brickyard, New Road, Gillingham, Dorset SP8 4XT) was prepared. The silica (50g) was dispersed in 5 moldm⁻³ nitric acid to clean it and the mixture filtered. The silica filter cake was washed with demineralised water, dried and calcined in air at 500°C for 2 hours. The Pt salt, Pt(NH₃)₄(OH)₂ (28.87g), was placed in a beaker and sufficient demineralised water added to

produce 100g of solution. The Pt salt solution was added to the calcined silica and the mixture stirred for 5 minutes to allow the silica to adsorb the Pt salt solution. The resultant thick slurry was dried overnight at 60°C in a vacuum oven. It was then calcined in air at 300°C for 2 hours. The resultant 5.6 wt% Pt catalyst supported on silica showed a high level of Pt dispersion on the silica support as shown by a CO metal area of 111m²/gPt and in transmission electron micrographs 1 to 3nm Pt particles, with some clustering forming collections of Pt particles 25nm across.

Preparation of Mixed Amorphous Silica Substrate

10 A mixture of chopped silica fibres (0.08g of type QC9/33 - 20mm from Quartz et Silice BP 521-77794 Nemours, Cedex, France and 0.47g of Q fibre type 104 from Johns Manville, Insulation Group, PO Box 5108, Denver, CO, USA) were dispersed with mixing in water (3000cm³). A non-woven matrix was fabricated from the resulting mixture in a single-step process, based on the principles of paper-making technology, at a sheet size of 15 855cm² (33cm diameter) in a sheet former (design based on standard SCA sheet former from AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden). The fibre sheet was removed from the wire and air dried at 150°C. The non-woven sheet was sprayed with Nafion[®] 1100 EW solution (5wt% solution in lower aliphatic alcohols from Solution Technologies Inc., Mendenhall, PA 19357, USA) to bind the fibres together. This gives a dry Nafion loading of 20 0.29mg/cm².

Formation of Composite Membrane

The mixed amorphous silica substrate was first cast with aqueous Nafion[®] solution (50g, 5wt% solution), prepared as described in patent specification numbers EPA 731 520 and 25 US 5,716,437, to produce, after drying, a substrate with a Nafion[®] layer applied to and partially embedded within one face of the substrate. Aqueous Nafion[®] solution (12g, 10.3wt%) was heated to 80°C and the 5.6 wt% Pt on silica catalyst (0.28g) added and the mixture silvered at medium speed for 10 minutes. The mixture was then sprayed onto the second face of the substrate to produce a thin layer of Pt catalyst and dried in the oven at 80°C. 30 This step was repeated until all of the mixture was sprayed onto the second face of the substrate, which was then dried at 150°C to produce a membrane. An identical membrane was

then prepared and the two membranes were laminated with the Pt catalyst face of each membrane facing each other at a temperature of 177°C and a pressure of 296psi for 6 minutes.

This produced a composite membrane of thickness 58µm (as measured by electron probe microanalysis (EPMA)) with a central region that contained the Pt on silica catalyst at a Pt loading of 0.05mg/cm² and with an overall Nafion[®] loading of 13.2mg/cm².

COMPARATIVE EXAMPLE 1

An identical pair of membranes were prepared from sheets of mixed amorphous silica substrate as detailed above, but without the addition of the Pt on silica catalyst. The two membranes were laminated at a temperature of 177°C and a pressure of 296psi for 6 minutes to produce a composite membrane of comparable thickness (55µm) and Nafion[®] loading (14.2mg/cm²) to the composite membrane containing the Pt on silica catalyst.

SINGLE CELL RESULTS

The membranes of Example 1 and Comparative Example 1 were formed into a membrane electrode assembly (MEA) using a carbon supported Pt/Ru catalyst containing anode (0.25mgPt/cm²) and a carbon supported platinum catalyst containing cathode (0.60mgPt/cm²). An evaluation of the MEA comprising the membrane was performed in a single cell operating at a temperature of 80°C with H₂/O₂ as reactants each at a pressure of 3 atmospheres absolute and reactant stoichiometries of 1.5 for H₂ and 10.0 for O₂.

A clear determination of the degree of H₂ cross-over from the anode to cathode of the MEA is provided by the open circuit voltage (OCV). The OCV is the cell potential when there is no net current flowing in the single cell and is determined by the difference between the cathode and anode potentials. Since hydrogen oxidation at the anode is extremely facile the anode potential is essentially constant in the MEAs examined herein and, consequently the OCV is a relative measure of the cathode potential. The cathode potential is a mixed potential which is determined by the electrode potentials for oxygen reduction ($\frac{1}{2}\text{O}_2 + 2\text{H}^+ + 2\text{e}^- = \text{H}_2\text{O}$), carbon corrosion ($\text{C} + \text{H}_2\text{O} = \text{CO}_2 + 2\text{H}^+ + 2\text{e}^-$) and hydrogen oxidation ($\text{H}_2 = 2\text{H}^+ + 2\text{e}^-$).

Since the change in the oxygen reduction and carbon corrosion potentials is negligible for the MEAs tested herein, any change in the OCV reflects a change in the concentration of hydrogen at the cathode i.e. reflects a change in the rate of hydrogen cross-over from anode to cathode through the membrane of the MEA.

5

Table 1 shows the OCV values of the MEAs based on the composite membrane of Example 1 at a platinum loading of 0.05mgP/cm² and the composite membrane of Comparative Example 1 that does not contain the Pt on silica catalyst. Also, shown for further comparison are the OCV values for identical MEAs based on commercially available Nafion[®] membranes (from E.I. DuPont de Nemours and Co. 22828 NC 87 Hwy. W. Fayetteville, NC 28306). The OCV of the MEA based on the composite membrane of Example 1 is significantly higher than either the OCV of the MEA based on the composite membrane of Comparative Example 1 that does not contain the Pt on silica catalyst or the OCV values of the MEAs based on the Nafion[®] membranes. This increase in OCV reflects a decrease in the hydrogen concentration at the cathode reflecting the reduction in the rate of hydrogen cross-over due to the recombination of the H₂ with O₂ at the Pt on silica catalyst located within the composite membrane. The extent of the reduction in hydrogen cross-over can be calculated from the change in the OCV by using the Nernst Equation which shows,

$$(OCV^2 - OCV^1) = RT/nF \ln (p_{H_2}^1 / p_{H_2}^2)$$

where p_{H₂} is the hydrogen pressure at the cathode. By using this equation the H₂ cross-over relative to Nafion[®] 117 can be calculated. As shown in Table 1 the rate of H₂ cross-over through the composite membrane of Example 1 is reduced by 85% compared to both Nafion[®] 117 and the composite membrane of Comparative Example 1, which does not contain the Pt on silica catalyst. This is despite the composite membrane of Example 1 being significantly thinner than Nafion[®] 117 in the fabricated MEAs (from EPMA measurements) as shown in Table 1. Further, as expected Nafion[®] 117 gives the lowest H₂ cross-over of the commercial membranes tested, since it is the thickest. This is shown in Table 1 by the H₂ cross-over of the commercial membranes relative to Nafion[®] 117, with all showing increased rates of H₂ cross-over with as expected, the progressively thinner Nafion[®] membranes showing progressively higher rates of H₂ cross-over relative to Nafion[®] 117. This highlights just how effective the composite membrane of Example 1 is at reducing H₂ cross-over by recombining

the H₂ with O₂ to produce water. This allows thinner membranes to be employed in the PEMFC, providing the benefits of both higher MEA performance and operation with reduced humidification, due to a higher water content in the membrane and the thinner membrane both promoting the rate of water back- diffusion from the cathode to anode.

5

Table 1

OCVs and reduction in H₂ cross-over due to the composite membrane containing the Pt on silica catalyst

Membrane	OCV / V	Membrane Thickness ^a / μm	H ₂ cross-over Relative to Nafion [®] 117 / %
Example 1	1.058	58	- 85
Comparative Example 1	1.029	55	0
Nafion [®] 112	1.005	40	+79
Nafion [®] 1135	1.018	75	+52
Nafion [®] 115	1.027	100	+12
Nafion [®] 117	1.029	150	0

10

^a From EPMA measurements

CLAIMS

1. A process for preparing a solid polymer electrolyte membrane comprising an ion-conducting polymer, a catalyst and a high surface area support material, which process
5 comprises:
 - (a) associating the catalyst with the support material to form a catalysed support;
and
 - (b) combining the catalysed support with an ion-conducting polymer composition.
- 10 2. A process according to claim 1, which comprises, as step (b), combining the catalysed support with an ion-conducting polymer in a liquid medium that is aqueous-based and is essentially free from organic solvents.
- 15 3. A process according to claim 1 or claim 2, wherein the catalyst comprises one or more precious metals, or combinations thereof, and/or other transition group metals.
4. A process according to any preceding claim, wherein the catalyst comprises platinum.
- 20 5. A process according to any preceding claim, wherein the catalyst is deposited onto the support material to a loading of between 0.01 to 50.0% by weight of the total catalysed support.
- 25 6. A process according to claim 5, wherein the catalyst is deposited onto the support material at a loading of from 1 to 25 wt% of the total catalysed support.
7. A process according to claim 6, wherein the catalyst is deposited onto the support material at a loading of from 1 to 10 wt% of the total catalysed support.
- 30 8. A process according to any preceding claim, wherein the amount of catalysed support incorporated into the membrane is such that the metal loading is lower than 0.1mg/cm².

9. A process according to claim 8 wherein the amount of catalysed support incorporated into the membrane is such that the metal loading is lower than $0.05\text{mg}/\text{cm}^2$.
- 5 10. A process according to claim 9 wherein the amount of catalysed support incorporated into the membrane is such that the metal loading is lower than $0.03\text{mg}/\text{cm}^2$.
11. A process according to any preceding claim, wherein the high surface support material is non-electrically conducting.
- 10 12. A process according to any preceding claim, wherein the high surface area support material is selected from silica, titania, alumina, zirconium oxides, zirconium silicates, tungsten oxides, tin oxides and zeolites.
13. A process according to any preceding claim, wherein the support material is in the
15 form of fibres.
14. A process according to any one of claims 1 to 12, wherein the support material is in the form of particles with a mean particle size in the range of from $0.001\mu\text{m}$ to $10\mu\text{m}$.
- 20 15. A process according to claim 14, wherein the mean particle size is in the range of from $0.01\mu\text{m}$ to $5\mu\text{m}$.
16. A process according to any preceding claim, wherein the ion-conducting polymer composition is an essentially aqueous solution of a perfluorinated co-polymer with
25 ion-exchange groups.
17. A process according to any preceding claim, wherein the catalysed support, in particle or fibre form, is added directly to a solution of the ion-conducting polymer electrolyte.
- 30 18. A process according to any of claims 1 to 11 or 13 to 15, wherein the catalysed support is in particle form and is applied as a binder to form a fibre network to which the ion-

conducting polymer is subsequently applied to produce the membrane.

19. A process according to any of claims 1 to 13 or 16, wherein the catalysed support is in fibre form and itself is formed into a fibre network which is thereafter bound with a binder, and the ion-conducting polymer is subsequently applied to produce the membrane.
20. A membrane prepared by a process according to any preceding claim.
21. A membrane electrode assembly comprising a membrane prepared by a process according to any one of claims 1 to 19.
22. A fuel cell comprising a membrane prepared by a process according to any one of claims 1 to 19.
23. A fuel cell comprising a membrane electrode assembly according to claim 21.

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE,

the specification of which is attached hereto unless the following box is checked:



was filed on October 4, 1999 as

United States Application Number or PCT International Application Number PCT/GB99/03269

and was amended by Preliminary Amendment filed along with the application (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Not Claimed

9822576.6

Great Britain

16 October 1998

(Number)

(Country)

(Day/Month/Year Filed)

☐

(Number)

(Country)

(Day/Month/Year Filed)

☐

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Number)

(Filing Date)

(Status - patented, pending, abandoned)

(Application Number)

(Filing Date)

(Status - patented, pending, abandoned)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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